Finite Temperature Magnetic Properties of Rare Earth Iron Garnets

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Background

Magnetic garnets as insulators are very important materials which have been studied extensively since the mid-twentieth century with promising outlooks in applications over many different areas of research. More specifically, Rare Earth - Iron Garnets (RIGs) of the formula $R_3Fe_5O_{12}$ (Fig. 1) are especially promising because of their low Gilbert damping, high Curie temperature, and tunable magnetic properties [1]. They have been studied experimentally and theoretically in the past, but in order to fully understand the magnetic properties of these garnets, a computational atomistic approach should be applied. This will give, e.g., more detailed information about how exactly these structure respond to various magnetic conditions, and therefore tell us how they might be used in applications such as spintronics, thermoelectric generation, and information storage [2-4].

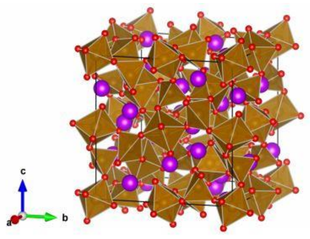

Figure 1: $R_3Fe_5O_{12}$ structure: iron in brown, oxygen in red, and rare earth in purple [1]

Magnetic Properties

RIGs have been found to have two different types of Fe^{3+} ions: those that form the tetrahedral molecular geometry and those that form the octahedral geometry (Fig. 2.) These different ions have been found to have opposite magnetic moments of differing magnitude, meaning the garnets are *ferrimagnetic*. In addition to this, the rare-earth ions have been found to have a drastic increase in their magnetic moment below a specific temperature, called the compensation temperature, T_M. This moment is in the same direction as that of the octahedral iron, and, at low enough temperatures, stronger in magnitude than both iron ions [1]. Using the equation:

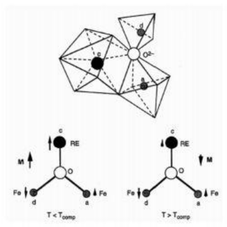

Figure 2: Diagram of the bond angles of the octahedral and tetrahedral irons [1]

$$M_{\text{total}} = M_{\text{Fe}_{\text{Tet}}} - M_{\text{Fe}_{Oct}} - M_{\text{R}}$$

one can find the total magnetic moment of the RIG. As the moments of both iron ions and the rare-earth ions can be strongly temperature dependent, as the temperature decreases to 0 K, one can find a compensation temperature at which the RIG has zero net magnetic moment [2]. This temperature has been found to coincide with the relatively low temperature at which the rare-earth elements begin to have drastic changes in magnetic moment [1]. It must be also noted that the oxygen atoms in the supercell have no contribution to the magnetic properties as far as the extent of this paper is concerned.

Previous Work

Because of the relatively large size of these RIGs (160 atoms per unit cell), few first-principle calculations were carried out on them to determine properties until reference [1].

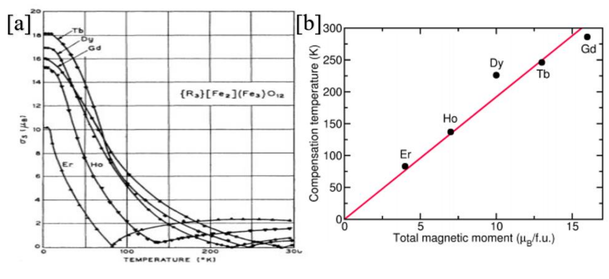

Figure 3: [a] Experimentally found values for the net magnetic moment of various RIGs as a function of temperature [2]; [b] Total magnetic moment *versus* compensation temperature for various RIGs [1].

Experimental results (Figure 3a) along with first-principle calculations (Figure 3b) have been carried out to investigate the relationship between the rare earth ion and the compensation temperature of the RIG.

Motivation

In this study, we further investigate the magnetic properties of a rare-earth garnet to a greater depth. We perform calculations to see the effects of multiple processes on the magnetic properties of the RIGs. These processes include zero field cooling and field cooling with varying levels of applied magnetic field. We hope to gain a significantly better understanding of the magnetic properties of RIGs, and we possibly look to uncover any new phenomena that may be possible with them.

Results

We see that, as the supercell is cooled from 800K to approaching 0K, it first reaches the Curie temperature around 600K. This is the point at which the moments of the tetrahedral and octahedral iron ions begin to be ordered, providing a finite value magnetization of the system. Next, the compensation temperature is reached at about 280K, which is the next point at which the net moment of the supercell reaches zero. At this point, the gadolinium ions begin to be ordered in moments quite rapidly, leading to overtaking and quickly dominant influence of the net magnetic moment of the supercell. By the time the temperature is approaching 0 K, the net moments of the gadolinium, octahedral iron, and tetrahedral irons are 21, 10, and -15 Bohr Magnetization per formula unit (μ_B/f.u.) respectively, leading to a net moment of 16 μ_B/f.u. The total size of the supercell used was 4 x 4 x 4 unit cells of 160 atoms each, totaling 10,240 atoms (6144 oxygen, 1024 octahedral iron, 1536 tetrahedral iron, and 1536 gadolinium.)

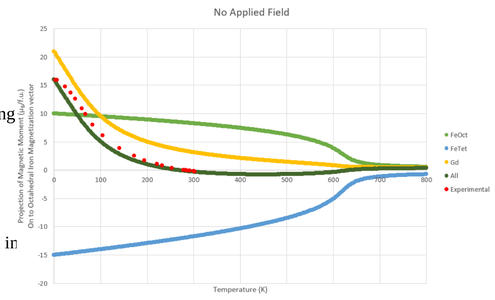

Figure 4: Magnetic moment of various elements in the supercell *versus* temperature

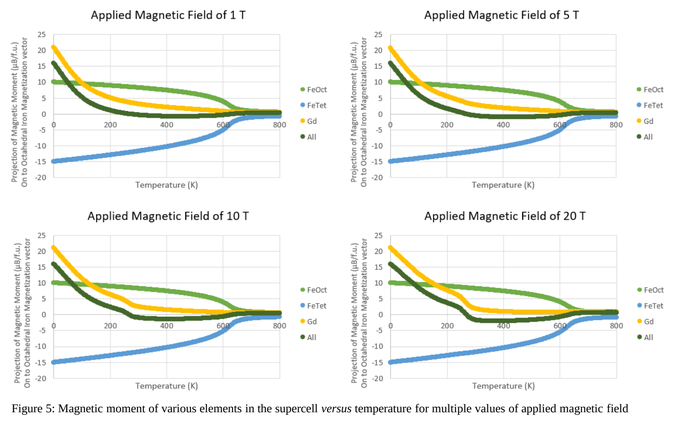

Figure 5: Magnetic moment of various elements in the supercell *versus* temperature for multiple values of applied magnetic field

Next, the system was looked at in the same manner as before, but, this time, a magnetic field was applied in the [001] direction. Figure 5 shows the net magnetic moment of the supercell *versus* temperature under different magnetic conditions. It then becomes apparent that the application of the field is greatly affecting the rate at which the gadolinium ions align once the compensation temperature is reached, while the external field has little effect on either of the irons. Both the compensation temperature and Curie temperature were also found to have no significant change in response to a change in the magnitude of the applied field. It must be noted that the direction of the applied magnetic field is of no importance. No matter the direction of the applied field, the same results are found because of our adopted collinear magnetism.

We then look at the specific heat capacity *versus* temperature for the system under different applied fields (Fig. 6). Note that the large at 583K is associated with the phase transition at the Curie temperature. As the applied magnetic field at the compensation temperature is increased, the peak at that point becomes significantly more defined. This would initially indicate that there is a phase transition of some kind at that temperature.

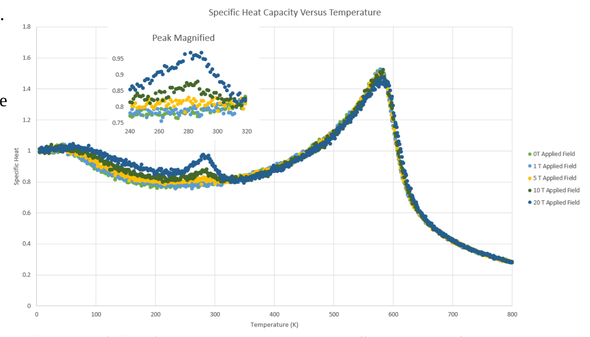

Figure 6: Specific Heat of the supercell *versus* temperature under different magnitudes of applied magnetic field

In order to ascertain that there is a phase transition, we next analyze the magnetization vectors of the supercell across all temperatures and all applied magnetic fields.

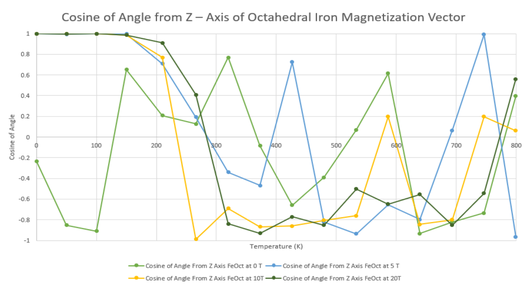

Figure 7: Cosine of the angle from the Z-axis of the unit vector in the direction of the net magnetization for octahedral iron. Lines are added between data points to more easily see the change from point to point.

Figure 7 plots the angle from the Z - axis of the octahedral iron magnetization vector as a function of temperature for different levels of applied field. At 0T applied field, the direction of the magnetization vector with respect to the Z - axis is seemingly random, as consistent with collinear magnetization. This is because of the lack of Zeeman effect - which states that the total magnetic moment should align with the direction of applied magnetic field. There is no applied magnetic field, so the net magnetic moment will point in some random direction. With an applied field, however, the Zeeman effect is demonstrated, and the total net magnetic moment is in the direction of the applied magnetic field ([001] direction). As can be seen in the graph, once the temperature hits the compensation temperature, the magnetization of the octahedral iron flips from along the negative Z - axis to strongly along the positive Z - axis. This change is more pronounced under an applied field of 10T or greater, which is also the level of applied field at which we better saw the peak in the specific heat *versus* temperature graph.

Conclusions

To summarize, what we have found about the change at this compensation temperature is that, above that temperature, the total magnetic moment of the system is aligned with the moment of the tetrahedral iron (opposite to the octahedral iron). This means, based on the Zeeman effect, the magnetization of the tetrahedral iron will tend to align with the applied magnetic field. Below the compensation temperature, however, the total magnetic moment is in the direction of the octahedral iron and gadolinium, and the magnetization of the tetrahedral iron prefers to be anti-parallel to that of gadolinium and tetrahedral iron. This results in the magnetization of the tetrahedral iron preferring to be aligned anti-parallel to the magnetic field below the compensation temperature. This finally means that there is indeed a phase change at finite applied magnetic field around the compensation temperature that is the reverting of the magnetization of *all* three types of ions in the system, as schematized in Figure 8.

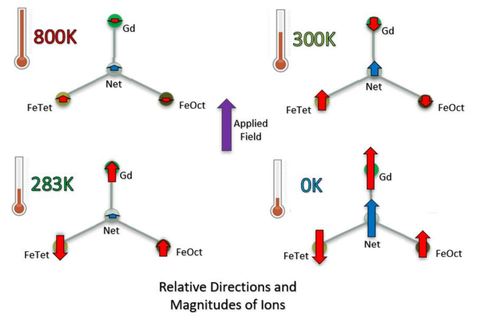

Figure 8: Depiction of the phase transition under applied magnetic field as temperature is decreased past the compensation temperature. The red arrows indicate the relative magnitude and direction of the moments of the ions.

References

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Methods

We begin by applying DFT using the Vienna *ab initio* simulation package (VASP) to get the magnetic parameters of the RIGs. We first write the spin Hamiltonian between two ions in the supercell as

$$\hat{H}_{\text{spin}} = \sum_{i<j} J_{ij}\vec{S}_i \cdot \vec{S}_j - c\sum_k \vec{B}\cdot\vec{S}_k$$

where the first term describes the collinear interactions between the ions, the second term describes the effect of an applied magnetic field on the spins, and *c* is a constant for unit conversion [5].

We can apply DFT as defined above on four ordered spin states to determine the J_{ij} parameters [5]. Those states are as follows:

	Spin 1	Spin 2
State 1	+1	+1
State 2	+1	-1
State 3	-1	+1
State 4	-1	-1

The application of DFT to each of these states give us an energy value which we can finally apply the following equation:

$$J_{ij}^{e} = \frac{E_1 + E_4 - E_2 - E_3}{4}$$

Which gives us the spin lattice coupling between any two ions in the supercell. It is an important note to make that we use collinear magnetism to extract the magnetic parameters, implying that, under no applied field, there is no energy preference to any direction of the total magnetic moment of the system.

From these electronic configurations, we re-scale the couplings and run various Monte-Carlo heat bath simulations on the data. Using the Heat Bath Monte-Carlo code [6], We can look at many different properties of the RIGs under many different conditions.

The instance of the heat bath code that we run allows us to look at the magnetic properties of the garnets under a variety of scenarios. We begin with the Monte-Carlo code by first running the same zero field cooling (ZFC) computations that have been done both experimentally [2] and computationally [1] before in order to adjust the aforementioned scaling factors until the calculations match the experimentally found results. From there, we look at the RIGs under field cooling. Field cooling (FC) is the experimental procedure of applying a magnetic field to the compound while at the same time cooling the material down, as opposed to the ZFC mentioned before. We use this procedure to see the effect of the application of a magnetic field on the magnetic moments of each individual atom, and then of course the net moment of the system.

The calculations were carried out on the University of Arkansas high performance computing cluster, Razor. All the data was then processed using Microsoft Excel, and all figures were made in either the modeling software Vesta [7] or in Microsoft Excel.

RIG Selection

This work was done with only the Gadolinium RIG, as it was seen to be the most promising of the possible rare earth elements due to its possession of the highest compensation temperature of all the RIGs [1]. This compensation temperature is relatively close to room temperature, which makes it an extremely promising material for applications such as moving domain walls at room temperature.

Acknowledgments

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